ELECTROKINETICS

II. – Development of Equations for Two-Phase Flow Through Single Capillaries

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ABSTRACT

This report is the second part of a study on the electro-kinetics of the flow of oil and water through petroleum reservoirs.

The development of electric charges at the oil-water inter-face is discussed and equations are presented for the streaming potential, electro-osmotic pressure, and electroviscous effect displayed by a single capillary filled with an annulus of wetting phase (water) and having a central core of immiscible nonwetting phase (oil).

Although it is unlikely that oil and water would flow through a porous medium following the theoretical pattern set forth here, it seems certain that some fraction of the flow is of this type and would modify considerably the measured electrokinetic effects.

INTRODUCTION

Although the effect of electrokinetics in retarding the flow of a single phase through capillaries and porous plugs has been treated theoretically and observed experimentally, no comparable work has yet been done on the simultaneous flow of two immiscible fluids. There seems little doubt that during such flow both fluids, under suitable conditions, undergo an electroviscous retardation; but it is also possible that, under other conditions, one of the phases may be accelerated so that the measured viscosity is less rather than greater than the bulk viscosity. Such decreased viscosity actually has been observed by Templeton (1954) and Templeton and Rushing (1956) during capillary flow of water and oil through micro-scopic capillaries.

Before it is possible to derive an equation for the electroviscous effect resulting from two-phase flow through a porous medium, it is necessary to develop a model for that flow. Generally such a model is built up from some combination of capillary tubes. Thus the simplest theoretical approach is to equate the porous medium to a single capillary of suitable length and radius for the purpose of describing its hydrodynamic behavior. The logical extension is to a combination of tubes of varying radii, arranged in parallel and series. Such a series model was used by Scott and Rose (1953) to modify the Yuster model and thus help explain the nondependence of relative permeability on viscosity ratio.

Network models (interconnecting networks of parallel and series elements) have been analyzed by Fatt (1956) who found them useful in predicting flow behavior; this work has been extended by Rose (1957). The use of network models for analyzing electrokinetics in porous media has been treated by Overbeek and Wijga (1946), Mazur and Overbeek (1951), and by Lorenz (1953a).

Wyllie and Gardner (1958) suggested that one might start with a bundle of capillary tubes, cut them into thin sections, and rearrange these sections at random laterally to produce a "pseudo-model."

As pointed out by Templeton, the first concern is to gain knowledge of the mechanism of the flow through individual capillaries.

Using a "bundle of capillary tubes" model, Brownell and Katz (1947) suggested that both phases flow through all the tubes, whereas Wyllie and Gardner suggested that each phase follows a separate path.

It is possible to combine these views (as I plan to do in another article) and to divide the flow into three regions: a) tubes flowing both wetting and nonwetting phase; b) tubes flowing only wetting phase; and c) the remainder of the tubes, wetted by an immobile film of wetting fluid and flowing the nonwetting phase.

The percentage of the total flow in each region can be determined by analysis of relative permeability curves carried out when the electrokinetic effect is negligible. From the contribution of each flow type to the total streaming potential, or volume flowed during electro-osmosis, one can then calculate the total streaming potential or electro-osmotic pressure to be expected, and thus also predict the electroviscous retardation to flow.

CHARGED OIL-WATER INTERFACE

There is much experimental evidence that charged interfaces develop between oil and water, but the distribution of charges at this interface is open to discussion. Thus Booth (1951), in deriving equations for the calculation of zeta potentials from the electrophoretic mobility of oil droplets dispersed in an aqueous medium, considers three possible distributions of charge: a) the charge is concentrated in a thin layer at the interface; b) the charge is distributed in uniform density throughout the droplet; c) the charge is distributed in an electric double layer developed in the oil phase.

The equations developed for distribution c reduce to those for distribution b when the double-layer thickness becomes greater than the radius of the droplet. One may consider that distribution c is the intermediate one and that the double layer can expand into uniform density, b, or compress into a thin interfacial layer, a. Thus if we picture the general charge distribution at the oil-water interface to be that of two diffuse double layers, we can illustrate it schematically as in figure 1.

Here, since the double diffuse double layer as a whole must be electrically neutral
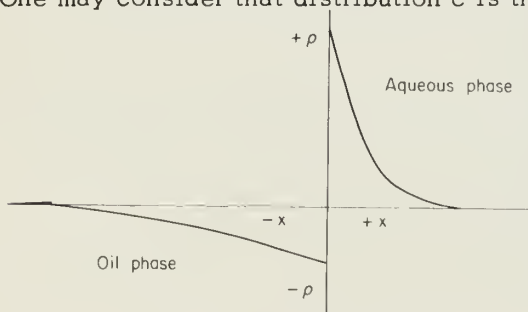
Fig. 1. – Schematic representation of the distribution of space charges in the oil and water phases

$$\int_{-\infty}^{0} \rho_1(x)dx + \int_{0}^{\infty} \rho_2(x)dx = 0 \qquad (1)$$

which will also describe the charge condition in the event of reduction to either a or b. ρ_1, ρ_2 = charge density in phase 1 and phase 2, respectively.

The double-layer thickness in either phase is generally given by the expression

$$\lambda = \sqrt{\frac{DD_okT}{2nz^2e^2}} \tag{2}$$

where

k = Boltzmann's constant
T = absolute temperature
n = number of dissociated molecules per m^3
z = valency of ions
e = charge of an electron
D = relative dielectric constant
D_o = absolute dielectric constant of a vacuum

However, for purposes of comparison between aqueous solutions and hydrocarbons the expression given by Klinkenberg and van der Minne (1958) is much to be preferred. This expression may be arrived at from (2) by a series of substitutions, thus

$$\Delta_m = \frac{kT}{6\pi\eta a}$$

where

Δ_m = coefficient of molecular diffusion
a = radius of an ion
η = viscosity of medium

and

$$v = \frac{F}{6\pi\eta a}$$

where

v = ionic velocity under the influence of an electric force, F
F = zeE (E is electric field strength)

It is also true that the current, I, flowing under the influence of a potential difference, E, in a liquid of conductivity, K, is

$$I = KE = 2nvze$$

Hence it follows that in (2) kT may be replaced by

$$\Delta_m 6\pi\eta a \quad \text{and} \quad 2nz^2e^2 = K\frac{Eze}{v} = K\frac{Eze}{I}\frac{6\pi\eta a}{zeE} = K6\pi\eta a$$

and so

$$\lambda = \sqrt{\frac{DD_o\Delta_m}{K}}$$

Klinkenberg and van der Minne point out that Δ_m may be assumed to have about the same value in hydrocarbon and water, because in the hydrocarbon the somewhat larger size of the organic ions will be compensated for by the lower viscosity of the medium.

Early experiments on the mobility of oil droplets dispersed in water were reported by Ellis (1912), Mooney (1924), and Limberg (1926). Some authors have reported their results in terms of mobilities (microns per second per volt per centi-

meter), and others as zeta potential
(millivolts). Although for large spherical
solid particles dispersed in water at 25°C
the zeta potential is numerically equal to
the mobility multiplied by 14.3, such a
simple relationship does not apply be-
tween the mobility and zeta potential of
oil droplets.

Fig. 2. - The flow model showing the dis-
tribution of the phases and of their
associated double layers

First there is the problem of the
charge distribution, which may be one of
the three possible distributions listed by
Booth, and, second, there is the fact that
the droplets are liquid, which causes them
to have a velocity different from that of
an equivalent solid particle. Booth suggests that the mobility of the equivalent
solid particle will be greater, but Jordan and Taylor (1952) suggest it will be less;
Jordan and Taylor's hypothesis is supported to some extent by experimental evi-
dence.

It would appear that the zeta potentials of paraffin oil droplets may approach
values of -86 mV in water and -151 in 0.0036M sodium oleate (Urbain and Jensen,
1936). Powney and Wood (1940) report mobilities of 4.35 μ/sec/v/cm for Nujol
droplets decreasing to about 2 at 0.005N. The mobility was considerably increased
when the droplets were dispersed in sodium oleate and sodium dodecyl sulphate
solutions. Newton (1930) had reported velocities of 3.9 in water with a conduc-
tivity of 4×10^{-6} ohm^{-1} cm^{-1}.

More recently Douglas (1950) found that dissolved sugar reduced the mobil-
ity of hydrocarbons in solutions of nonionic substances. He explained this as being
due to the preferential adsorption of the sugar, which reduced the ionic adsorption
and consequently the surface charge density.

Many workers have studied hydrocarbons other than paraffin. Taylor and
Wood (1957) found zeta potentials of -70 to -100 mV for decalin droplets in NaCl
solutions, and Carruthers (1938) attempted to correlate measured mobilities with
chemical composition using n-Octadecyl, n-Undecyl, n-Octyl, and $\Delta^{10,11}$n-Un-
decenyl alcohols and n-Octadecane and $\Delta^{1,2}$n-Octadecene all dispersed in solu-
tions 0.01N with respect to sodium ion. Douglas (1943) determined the mobility
of droplets of paraffin wax, dodecane, octadecane and $\Delta^{1,2}$octadecene, and of
mixtures of hexane-dodecane, cyclohexane-dodecane, benzene-dodecane and
decalin-dodecane, getting values of 1 - 1.5 μ/sec/v/cm in solutions kept 0.01N
in sodium ion at various pH's.

DEVELOPMENT OF EQUATIONS

This paper deals with the electrokinetics to be expected when two phases
are present in one capillary tube, and considers the flow through a single capillary
under the conditions suggested by Brownell and Katz (1947) and by Yuster (1951),
namely that the nonwetting phase (oil) should flow in a cylindrical portion of the
wetted (by water) capillary and concentric with it (fig. 2).

These authors assume that the velocities of the oil and aqueous phases are
equal at the interface during such flow, and Yuster gives the following expression
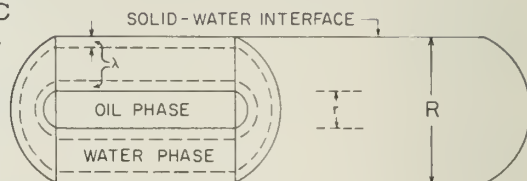for the velocity distribution in the oil phase

$$u_0 = \frac{P}{4L}\left[\frac{(R^2-r^2)}{\eta_w} + \frac{(r^2-a^2)}{\eta_o}\right]$$ (4)

where
u_0 = velocity of laminar layer of radius a
R = radius of capillary
r = radius of oil phase
η_w = viscosity of water phase
η_o = viscosity of oil phase
P = pressure drop across capillary
L = length of capillary

The average velocity of the oil phase is

$$v_0 = \frac{P}{8\eta_o Lr^2}\left[2(R^2r^2-r^4)\frac{\eta_o}{\eta_w} + r^4\right]$$ (5)

and the velocity (v_w) of any laminar layer (radius a) in the aqueous phase is

$$v_w = \frac{P}{4\eta_w L}\ [R^2-a^2]$$ (6)

Streaming Potential

Assume that both the capillary and the oil surfaces are negatively charged so that mobile positive counter ions are located in the aqueous phase. (The analysis will remain valid, with due attention to signs of the zeta potentials, in the event of other charge distributions.) As only one ionic solution is in contact with both the capillary wall and the oil surface, it is apparent that the double layers associated with both surfaces will be of the same thickness (λ). Then assuming, for simplicity in the derivation, that all the counter ions are located in a Helmholtz double layer, it is possible to derive an expression for the streaming potential caused by the movement of charges in the aqueous phase (Wood, 1946; Butler, 1940; Perrin, 1904).

The streaming potential arises partly from the transport of the positive ions of both the double layers in the aqueous solution, namely those associated with the capillary wall charge and those associated with the oil surface, and partly from transport of the negative charges associated with the oil itself. It is of course essential that the thickness of the aqueous phase be such that the double layers do not overlap and that the distribution of charges in the oil phase be known.

It is possible to suggest that the charges in the oil phase are located in a Helmholtz double layer at a distance (λ_o) of approximately

$$\lambda_o = \frac{D_o K}{D K_o}\cdot\lambda$$

from the oil-water interface (D_o and K_o are the dielectric constant and the conductivity of the oil, respectively).

However, the double-layer thickness in the hydrocarbon phase will be great, and, as we consider only capillaries of small diameter, it is more likely that the charge is homogeneously distributed throughout this phase as suggested by Klinkenberg and van der Minne (1958).

(1) Oil Phase

If electrokinetic measurements in the aqueous phase indicate a surface charge density of

$$-\int_o^\infty \rho_2(x)dx = -\sigma_2$$

on the oil phase, then the surface charge per unit length is $-2\pi r\sigma_2$; an expression that describes the charge per unit length of this phase whatever the charge distribution.

If the oil should move with an average velocity, v_o, the current of convection is

$$I_o = -2\pi r\sigma_2 v_o$$

and substituting the value of v_o from equation 5 gives

$$I_o = -\frac{\pi r_2 P}{4\eta_o r}\left[2(R^2r^2-r^4)\frac{\eta_o}{\eta_w}+r^4\right] \tag{7}$$

(2) Aqueous Phase

Assuming that the surface-charge density in the Helmholtz double layer associated with the capillary interface is σ_1 and that associated with the oil-water interface is σ_2, the current caused by the transport of these charges is given by

$$I_w = 2\pi R\sigma_1 v_1 + 2\pi r\sigma_2 v_2$$

where v_1 and v_2 are the velocities in the double layer associated with the capillary surface and the oil surface, respectively — that is, v_1 is the velocity of radius $R-\lambda$ and v_2 is the velocity of the annular layer of radius $r+\lambda$. Thus

$$v_1 = \frac{P}{4\eta_w}[R^2-(R-\lambda)^2], \quad v_2 = \frac{P}{4\eta_w}[R^2-(r+\lambda)^2]$$

and

$$I_w = \frac{\pi P}{2\eta_w}[(2R\lambda)R\sigma_1 + (R^2-r^2-2r\lambda)r\sigma_2] \tag{8}$$

(3) Total Streaming Potential

Because the conductivity of the oil phase (10^{-10} to 10^{-12}) is much smaller than that of the aqueous phase (10^{-3} to 10^{-6}) the current of conduction will be given by

$$I_s = K\pi(R^2 - r^2)E$$

where

$$K = \text{conductivity of the aqueous phase}$$
$$E = \text{streaming potential}$$

The total current carried in the streaming liquids is $I = I_w + I_o$, and at equilibrium $I = I_s$, and so it follows when

$$S_w = \frac{R^2 - r^2}{R^2} \quad \text{and} \quad S_o = \frac{r^2}{R^2}, \quad \text{that}$$

$$E = \frac{P\lambda}{\eta_w K S_w} \left\{ \sigma_1 - S_o[1 + \frac{\eta_w R}{2\eta_o \lambda} S_o^{\frac{1}{2}}] \sigma_2 \right\} \tag{9}$$

or putting

$$\sigma_1 = \frac{D\zeta_1}{4\pi\lambda}, \qquad \sigma_2 = \frac{D\zeta_2}{4\pi\lambda}$$

then

$$E = \frac{DP}{4\pi\eta_w K S_w} \left\{ \zeta_1 - S_o[1 + \frac{R\eta_w}{2\lambda\eta_o} S_o^{\frac{1}{2}}] \zeta_2 \right\} \tag{10}$$

ELECTRO-OSMOTIC PRESSURE

By considering the development of an electro-osmotic pressure in each phase of this system we can consider conditions that exist when there is no movement at the fluid-fluid boundary. Hence the problem in the aqueous phase is simply that of the electro-osmotic pressure developed in an annulus, and the problem in the oil phase is the electro-osmotic pressure produced in a capillary tube with a homogeneous charge distribution in the liquid.

(1) Aqueous Phase

Following the method of Porter (1920) we have: total charge in unit length of annulus of radii R and r is

$$2\pi R\sigma_1 + 2\pi r\sigma_2$$

and hence with an applied potential difference of E the electric force causing movement is

$$F_1 = 2\pi(R\sigma_1 + r\sigma_2)E$$

The frictional forces opposing flow are

$$F_2 = -2\pi R\eta_w dv/da + 2\pi r\eta_w dv/da$$

The velocity within the double layer varies linearly with distance from either wall and is zero at a=r and a=R, and so within the double layer extending from R to R-λ, dv/da = -v/λ, whereas in the double layer extending from r to r+λ, dv/da = v/λ. It follows that

$$F_2 = 2\pi(R+r)\eta_w v/\lambda$$

At equilibrium $F_1 = F_2$, and so it follows that

$$v = \frac{(R\sigma_1 + r\sigma_2)E\lambda}{(R+r)\eta_w}$$

or

$$v = \frac{DE(R\zeta_1 + r\zeta_2)}{4\pi(R+r)\eta_w}$$

or when V is the volume rate of flow

$$V = \frac{DE(R-r)(R\zeta_1 + r\zeta_2)}{4\eta_w} \tag{11}$$

In order to express this in terms of an electro-osmotic pressure, equation 11 should be equated with the equation for flow through an annulus (Lamb, 1932). Thus

$$\frac{DE(R-r)(R\zeta_1 + r\zeta_2)}{4\eta_w} = \frac{\pi P_w}{8\eta_w}(R^2 - r^2)(R^2 + r^2 - \frac{R^2 - r^2}{\ln R/r})$$

and so

$$P_w = \frac{2DE(\zeta_1 + S_o^{\frac{1}{2}}\zeta_2)}{\pi R^2(1 + S_o^{\frac{1}{2}})(1 + S_o - \frac{S_w}{\ln S_o^{-\frac{1}{2}}})} \tag{12}$$

(2) Oil Phase

Total charge per unit length is $-2\pi r\sigma_2$, and with an applied field strength of E the electric force causing motion is

$$-2\pi r\sigma_2 E$$

The viscous force opposing motion is

$$2\pi r \cdot Pr/2\eta \cdot \eta$$

and so

$$P_o = -2E\sigma_2/r$$

or, expressing $-\sigma_2$ in terms of the zeta potential measured in the aqueous phase

$$P_o = -\frac{D\zeta_2 E}{2\pi r \lambda} \tag{13}$$

The negative sign indicates that the motion in the oil phase will be in a direction opposite to that in the aqueous phase with the assumed charge distribution.

(3) Total Electro-Osmotic Pressure

The aqueous phase electro-osmotic pressure (P_w) as derived acts over an area of $\pi R^2 S_w$, and the oil phase pressure (P_o) as derived acts over an area of $\pi R^2 S_o$. The total electro-osmotic pressure acting simultaneously over the area πR^2 is therefore
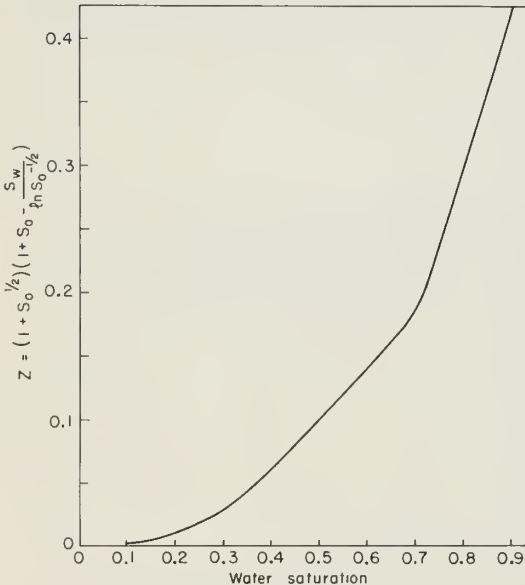
$$P = P_w S_w + P_o S_o$$

Fig. 3. – Water saturation against

$$Z = (1 + S_o^{\frac{1}{2}})(1 + S_o - \frac{S_w}{\ln S_o^{-\frac{1}{2}}})$$

Substituting from the expressions (12) and (13) gives

$$P = \frac{2DE\left\{S_w\zeta_1 + S_o^{\frac{1}{2}}(S_w - \frac{RZ}{4\lambda})\zeta_2\right\}}{\pi R^2 Z} \tag{14}$$

$Z = (1 + S_o^{\frac{1}{2}})(1 + S_o - \frac{S_w}{\ln S_o^{-\frac{1}{2}}})$ and is plotted in figure 3.

ELECTROVISCOUS EFFECT

In considering the electroviscous effect that arises out of this streaming potential and electro-osmosis, one must decide whether it is possible to test the effect microscopically or macroscopically. For example, Lorenz (1953b) in testing the effect during the flow of acetone through quartz plugs shorted out the ends of the plug and judged the existence of the effect from the differences in shorted and unshorted flow. He therefore, at least implicitly, considered that the effect was macroscopic in that he ignored the possibility that backflow might tend to occur in the liquid during its transport between the electrodes, and assumed that a potential difference must exist for the effect to be seen.

On the other hand, if certain viscosity increases in suspension flow in a concentric cylinder rotational viscometer are ascribed to the effect of electro-viscosity, one is implicitly supposing that the effect is microscopic and that it affects the particle at the instant of its displacement because in this case there is a closed circuit and no current flow.

The relevance of these concepts to the two-phase problem is that we could consider that each phase is reacted on independently by the streaming potential or, conversely, that both are reacted on together and to the same extent. In the first case we can sometimes predict an increase and sometimes a decrease for each phase, depending on such factors as saturation, viscosity ratio, and relative values of the surface charges. In the second, the electroviscous effect would always increase viscosity in the same ratio for each phase.

An expression for the electroviscous effect can be derived by considering the volume of electro-osmotic backflow, resulting from the streaming potential, that must be subtracted from flow caused by the pressure difference imposed on a tube. Or one can consider that the electro-osmotic back pressure reduces the pressure causing flow, a simpler idea to apply in the present case, although it is not to be supposed that these back pressures have any physical significance.

An expression for the electroviscous effect may be simply derived (Street, 1959) by finding a value for the electro-osmotic back pressure (P_1) that arises from the streaming potential and subtracting it from the applied pressure (P) to get the pressure (P_2) actually causing flow. Then

$$P_2 = P - P_1$$

If the liquid is flowing through a capillary tube of radius R and length L with a velocity v, its apparent viscosity (η_a) has a value of

$$\eta_a = \frac{P\pi R^2}{8Lv}$$

while the true viscosity (η) has a value of

$$\eta = \frac{P_2\pi R^2}{8Lv}$$

That is

$$\eta_a/\eta = P/P_2 \tag{15}$$

For both the aqueous and oil phases, P_2 is given by rearrangement of equation 10 as

$$P_2 = \frac{4\pi \eta_w K S_w E}{D\left\{\zeta_1 - S_o[1 + \frac{R\eta_w}{2\lambda\eta_o}S_o^{\frac{1}{2}}]\,\zeta_2\right\}}$$

The streaming potential E gives rise to an electro-osmotic back pressure (P_1) that has the value of P_w by equation 12 for the aqueous phase and of P_o by equation 13 for the oil phase.

Combining equation 15 with these values for the electro-osmotic pressure gives

$$\frac{\eta_{aw}}{\eta_w} = 1 + \frac{D^2(\zeta_1 + S_o^{\frac{1}{2}}\zeta_2)\left\{\zeta_1 - S_o[1 + \frac{R\eta_w}{2\lambda\eta_o}S_o^{\frac{1}{2}}]\zeta_2\right\}}{2\pi^2 R^2 \eta_w K Z} \tag{16}$$

$$\frac{\eta_{ao}}{\eta_o} = 1 - \frac{D^2\zeta_2\left\{\zeta_1 - S_o[1 + \frac{R\eta_w}{2\lambda\eta_o}S_o^{\frac{1}{2}}]\zeta_2\right\}}{8\pi^2 R\lambda\eta_w K S_w} \tag{17}$$

where

η_{aw}, η_{ao} = apparent viscosity of water and oil phases, respectively.

If the less likely macroscopic approach is used, then

$$P_1 = \frac{2DE\left\{S_w\zeta_1 + S_o^{\frac{1}{2}}(S_w - \frac{RZ}{2\lambda})\zeta_2\right\}}{\pi R^2 Z}$$

and so

$$\frac{\eta_a}{\eta} = 1 + \frac{2D^2[S_w\zeta_2 + S_o^{\frac{1}{2}}(S_w - \frac{RZ}{2\lambda})\zeta_2][\zeta_1 - S_o(1 + \frac{R\eta_w}{2\lambda\eta_o}S_o^{\frac{1}{2}})\zeta_2]}{\pi^2 R^2 Z\eta_w K S_w} \tag{18}$$

DISCUSSION

I do not believe that two immiscible fluids will actually flow through a porous medium according to this simple (and unstable) pattern. However, it seems certain that some fraction of the flow through porous media is of this type, and that even a small fraction of such flow would modify considerably the measured electro-kinetic effects.

It is probably impossible to test the equations in capillary flow experiments except at very low wetting-phase saturations, and then of course care must be taken to make sure that the double layers developed in the aqueous phase do not overlap, as has already been discussed (Street, 1959). The best test of the equations would appear to lie in experiments conducted with porous media for which the fraction of the various flow types has been determined.

The equations for streaming potential, electro-osmotic pressure, and electro-viscosity for porous media supposedly made up of bundles of capillary tubes of equal and varying radii will be presented in a future publication.

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